

# REPORT DOCUMENTATION PAGE

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14. ABSTRACT  Nuclear isomers can store tremendous amounts of energy for long times and the prospects for a controlled "clean" release of this energy for applications have motivated considerable research. Significant insight into nuclear structure also results from studies of reactions that may cause a triggered energy release. The primary emphasis has been on reactions induced by externally-produced photons. These studies comprise a very narrow sub-field of nuclear physics that has often been poorly connected with the broader results of traditional investigations. This report details efforts under this grant to provide improved measurements of trigger processes for several isomers and to enhance the basic foundation of knowledge connecting triggering studies with the body of nuclear physics. The significance of the work is reflected by fifteen publications, including a result recognized by the American Physical Society as a top story in physics during 2000.					
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# **NOTE**

This Final Technical Report for F49620-99-1-0263 covers the period of performance of May 1, 1999 – November 30, 2001. As such, introductory and background material reflects the state-of-the-art in the field at the end of 2001.

## INTRODUCTION

Long-lived excited states, or isomers, existing in many atomic nuclei are nature's nuclear batteries [1]. As such, isomers can store tremendous amounts of energy on an atom-by-atom basis and materials can be created that contain many such atoms. Concentrated into a solid, one gram of an isomeric material can store up to 1.3 gigaJoules, more energy than that contained in 100,000 grams of chemicals like gasoline or high explosives, for decades. One of the most attractive aspects is that the stored energy is often released from isomers entirely in the form of high-energy electromagnetic radiation. Since the emission of gamma rays allows the nucleus to de-excite without changing its internal composition, there is no direct transmutation of the nucleus with production of radioactive by-products by the triggered release. Of course, some isotopes do not have stable ground states and this would, following a triggered event, leave residual radioactivity. The ground-state decays themselves could provide a source of additional energy.

A number of possible applications [2] have been suggested for isomers, with a recent concentration on those that would benefit from a burst of gamma radiation, triggered upon demand and leaving a relatively "clean" residue. Possible concepts include:

- ◆ Delivery of therapeutic radiation in bursts upon demand directly to the site of tumors,
- ◆ Sterilization of biohazards or destruction of chemicals using bursts of radiation,
- ◆ Production of power for space and surveillance applications, and
- ◆ Creation of a laser that produces directional beams of coherent gamma rays; a *gamma-ray laser*.

For some applications, the ionizing quality of gamma rays would be most important, for example in damaging unwanted cells or other materials. The technological challenges are greatest in attempting to create a gamma-ray laser [3], but the result could be a device capable of manipulating matter at the smallest possible scales and delivering energy over great distances.

The promise of such applications, and others presently unforeseen, has motivated considerable research into why isomers exist, how they can be produced and how a release of the stored energy can be triggered. Of course, there is also a great deal of interest motivated purely on the basis of the insight into nuclear structure that isomers provide. Work conducted under this award has followed a broad approach to the study of triggered gamma emission from isomeric materials. In addition to experiments conducted and analyzed that directly address triggering for several isomers, other efforts were made to develop a base of fundamental knowledge of photon interactions with nuclei. Since examinations of triggering comprise a very focused area of research, often neglected by the mainstream of nuclear physics, this foundation is extremely important in connecting this area to more traditional and better established fields. It is clear from the

published literature that work conducted under this grant has been unsurpassed in the growing maturation of this field of study.

## TECHNICAL BACKGROUND

### Isomers

There is a key difference between the energy stored and released from isomers and that occurring in traditional fission and fusion reactions. In the latter, the energy is stored in the binding together of the constituent particles within a nucleus, and is released by breaking a large nucleus into smaller ones or fusing two small nuclei into a larger one. Either way there is a certainty of radioactive by-products. In isomers, the extra energy is stored in additional motion of the constituent nucleons within the nucleus. A release of isomer energy in the form of gamma rays simply reflects a reduction in the motion of those nucleons. For this reason, it has been said that isomeric energy is more kinetic than nuclear.<sup>1</sup>

Generally, energy can be stored within nuclei in the motion of the constituent particles or of the nucleus as a whole. The energies and angular momenta of single-particle states intrinsic to a nucleus are governed by the typical rules of quantum mechanics. One particularly important aspect is that most of the isotopes of interest for isomeric materials contain nuclei whose shapes are deformed from spherical. Usually the nuclear shape is prolate, somewhat like a rugby ball, having a unique body-centered axis. It is then possible to define the projection of the angular momentum vector associated with any energy level upon that axis. The quantum number associated with this projection is called  $K$  and assumes the primary role in understanding why some excited nuclear states can store such large amounts of energy for very long times [1].

Quantum mechanics dictates that for a nuclear state of specific angular momentum quantum  $J$ , the magnitude of the angular momentum vector is given by  $\sqrt{J(J+1)}\hbar$  and the magnitude of the projection (when it can be properly defined) is  $J\hbar \equiv K\hbar$ . If the individual motions of the constituent nucleons give rise, as occasionally happens, to an intrinsic state of large angular momentum, then that vector is oriented near the deformation axis (so-called *deformation aligned*). For example, a  $J=16$  intrinsic state of the nuclide  $^{178}\text{Hf}$  lies at an excitation energy of 2.445 MeV above the ground state. Therefore, this state is characterized by  $K=16$ , showing strong alignment of the angular momentum vector with the body axis of the nucleus. Electromagnetic decay of this state requires an energy-releasing transition to some lower-lying level, initiating a cascade that eventually reaches the ground state. It is known that only two intrinsic excited states are found lower than the  $J=K=16$  level, one with  $J=K=6$  and the other with  $J=K=8$ . The stable ground state is characterized by  $J=K=0$ . Thus, in all instances an electromagnetic decay transition from the 2.445-MeV level to other intrinsic states would require an extremely large multipolarity,  $L = \Delta J$ , suggesting a very low transition probability and a long lifetime.

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<sup>1</sup> Phrase suggested by J. D. Corey, Sandia National Laboratories.

Of course, quantum mechanics offers other possibilities for energy (and angular momentum) within a nucleus. With well-defined symmetry for a nucleus, its rotation as a whole is allowed. Collective rotation is characterized by a progression of states whose energy and angular momentum follow the pattern typically seen in diatomic molecules. Each intrinsic state, starting with the ground state, provides some base amount of energy and angular momentum to which one or more quanta of rotational energy and angular momentum may be added. A 'family' of states associated with the same internal motion of nucleons (corresponding to an intrinsic state) is referred to as a band with the intrinsic state being the *bandhead*. The most important feature is that collective rotation is only allowed by quantum mechanics around an axis that is perpendicular to the symmetry

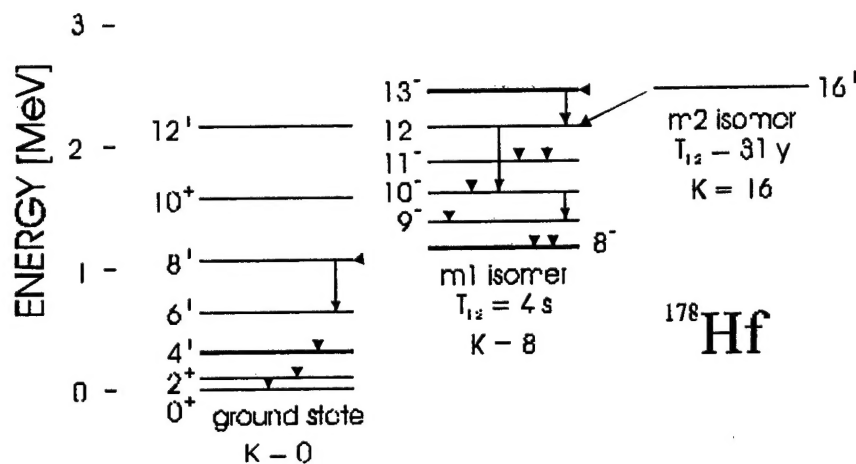


Figure 1: Schematic energy-level diagram showing known states of  $^{178}\text{Hf}^{m2}$  that participate in the natural decay of the 31-year isomer. The arrows indicate transitions that are part of the spontaneous decay cascade, which is 'stalled' as it reaches the 4-s m1 isomer.

(deformation) axis of the nucleus. Thus, more and more rotation, provided the shape of the nucleus is not too disturbed, increases the energy and total angular momentum associated with nuclear states, but does not change the projection of angular momentum on the symmetry axis. All members of the  $J=K=0$  ground state band (g.s.b.) in  $^{178}\text{Hf}$  have  $K=0$  up to at least  $J=20$ , reflecting that the angular momentum vectors of such levels are oriented along the rotation axis (so-called *rotation-aligned*).

Figure 1 shows a number of known energy levels of the isotope  $^{178}\text{Hf}$  with bands separated visually. Many more levels are known, of course. Due to the presence of these rotational bands, there are states to which the 2.445-MeV,  $K=16$  level can decay by modest multipolarity transitions. One example is that to the  $J=13$  member of the  $K=8$  band. As an electric octupole (E3) transition, it would cause the 2.445-MeV state to have a modest lifetime. There is, however, the additional problem of the required change in orientation of the angular momentum vector, reflected by the change in  $K$ .

The 2.445-MeV state is strongly deformation aligned, with  $K = 16$ . The state to which it could decay electromagnetically with the greatest probability (the aforementioned  $J = 13$  state) corresponds to only  $K = 8$ , due to the intrinsic bandhead with  $J = K = 8$ . Thus a transition to the  $J = 13$  state requires not only a reasonable change of  $\Delta J = L = 3$  in the magnitude of angular momentum of the nucleus, but an even greater change in the orientation. The total  $\Delta K = 8$  means that there is an excess orientation change of  $v = \Delta K - L = 5$ . Empirically, electromagnetic transition rates are reduced (inhibited) by such orientation changes according to a factor of about  $f^v$  with  $f \sim 10 - 100$  being typical for nuclei near  $A \sim 180$ ; for  $^{178}\text{Hf}$  isomers,  $f \sim 100$ . Thus, the 2.445-MeV state in  $^{178}\text{Hf}$  cannot decay even as rapidly as might be expected simply for an E3 transition, leading to a half-life of 31 years. This is an excellent example of a *K isomer*, storing an energy equivalent to 1.2 GJ/gram for a quite long time. Due to its high excitation (storage) energy, much research has focused recently on this isomer, referred to as  $^{178\text{m}2}\text{Hf}$ . There are many other examples of long-lived isomers and, in fact, the archetype is the  $10^{15}$ -year level of  $^{180}\text{Ta}$ .

### Triggering

The ability to trigger the emission of gamma rays from an isomer like  $^{178\text{m}2}\text{Hf}$  is the key to utilizing these high-energy density materials for most applications. Scientists have long sought to control the energy stored with the nucleus, but only since the invention of the laser in 1961 have there been any real concepts for triggering bursts of gamma rays without production of radioactive by-products. For much of that time the concentration was specifically aimed at finding a way to produce a gamma-ray laser, a task that has not yet been accomplished due to the conflicting demands of intersecting branches of physics [3]. Nevertheless, a large number of potential trigger mechanisms have been proposed and investigated both theoretically and, where possible, experimentally. The primary mechanism of interest has been called triggered (or induced) gamma emission and requires the irradiation of isomeric materials with x rays to initiate the release of energy from the isomers (see the review of Ref. [4]). Of course, for practical purposes it seems that the most attractive trigger events would require the smallest possible energy for the trigger in comparison with the amount of energy contained in the particular isomer.

The trigger mechanism depicted in Fig. 2 is a two-step process. An externally-produced x-ray photon incident upon the isomeric material is absorbed by a nucleus that is already in a long-lived metastable state. This absorption serves to excite the nucleus further into a level that must serve to mediate the trigger process, and is therefore referred to as an *intermediate state* or *trigger level*. For the absorption cross section to be large,

<sup>2</sup> The 'm2' indicates that this state is the second metastable level above the ground state, a somewhat arbitrary designation. The intrinsic state having  $J = K = 8$  shown in Fig. 1 has a half-life of 4 s and is thus referred to as the 'm1,' or first metastable. The other excited intrinsic state mentioned in the text is not shown in the figure. It lies slightly higher in energy than the m1 level, has  $J = K = 6$  and a half-life of 78 ns. Although this duration makes the half-life of the state measurable, it is not typically designated by the 'm' superscript.

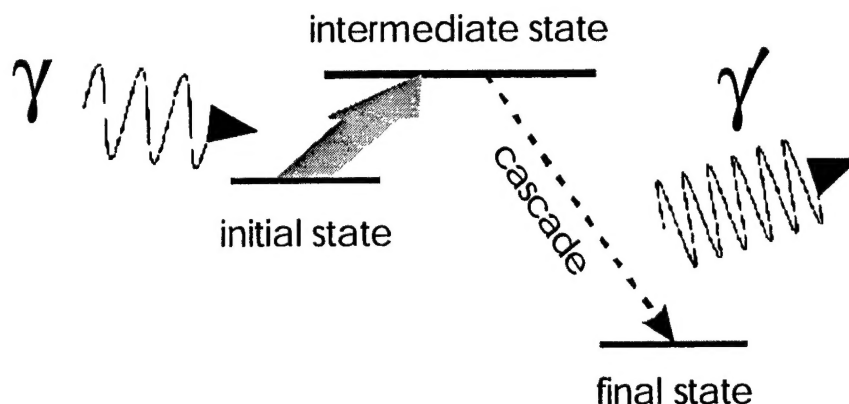


Figure 2: Schematic depicting the two-step process of triggered gamma emission. The heavy upward arrow shows excitation of the nucleus, by absorption of an incident photon, from the initial isomeric level to a higher-lying intermediate state. The intermediate then decays with some branch, shown by the dashed arrow, cascading to reach the final ground state. Energy released in this process is emitted by the set of gamma rays represented by  $\gamma$ .

leading to an efficient trigger reaction, the angular momentum of the intermediate state should be similar to that of the isomeric state. The greatest efficiency would likely result from dipole or quadrupole transitions.

In any case, the intermediate state may need to possess a strongly mixed wavefunction, containing components characterized by widely differing  $K$  values, in order to be excited from the high- $K$  isomer yet still be able to decay to low- $K$  members of the ground-state band. This mixing can occur due to near coincidences of nuclear states that lie in different bands and thus have different  $K$  values; nearly coincident states of varied  $K$  have recently been experimentally observed [5] in  $^{181}\text{Re}$ , but near a 12- $\mu\text{s}$  isomer, too short to be of value for energy storage. Also, if the nuclear interactions for a particular state disturb the axial symmetry of the nuclear shape, such as making it take a triaxial configuration, this would produce a strongly  $K$ -mixed wavefunction. Such an occurrence has been detected in  $^{186}\text{Os}$  [6].

Triggered gamma emission using x rays has been demonstrated conclusively for the isomer of  $^{180}\text{Ta}$  [7-12] and suggested for  $^{178\text{m}2}\text{Hf}$  [13-18]. The isomer  $^{180\text{m}}\text{Ta}$  is nature's only 'free' metastable state, since its halflife of more than  $10^{15}$  years has insured its survival into the present after its production in stars. The ground state of  $^{180}\text{Ta}$  is unstable, having a halflife of 8.2 hours, so all naturally-occurring nuclei of this isotope are in the isomeric state. Unfortunately for applications, that isomer lies only 75 keV above the ground state and it has been shown experimentally that x rays of at least 1 MeV [9] are required to trigger the release of the stored energy. The greatest probability for triggering is for x rays of 2.8 MeV [8, 9]. While unsuitable for applications, this and



other studies pointed the way [19, 20] to the possibility of triggering the  $^{178m2}\text{Hf}$  isomer, which lies at 2.445 MeV, through an intermediate state expected to lie below 2.8 MeV.

In 1998 simple experiments were performed [13, 14] in which a dental-style x-ray device was used to irradiate a sample containing  $6 \times 10^{14}$   $^{178}\text{Hf}$  nuclei in the 31-year-lived state. It was reported that the numbers of gamma rays emitted by some transitions in the spontaneous decay cascade of  $^{178m2}\text{Hf}$  were increased slightly during the irradiations, while the numbers of gamma rays emitted from a radioactive contaminant,  $^{172}\text{Hf}$ , did not change. The low energy of the accelerating potential, 90 kV, implied that triggering required x rays of less energy and that the process was quite efficient due to the ability to observe even a small enhancement of the gamma-ray lines with such a weak x-ray source. The large magnitude of the effect was a cause for concern in comparison with established nuclear systematics [21-24]. Unfortunately, relatively few experiments have been performed at such low energies - in general, the extensive literature of photonuclear reaction studies is confined to higher energies, greater than about 1 MeV and to excitation rather than triggering of isomers. To some degree this has been due to a lack of interest by the mainstream nuclear physics community in low-energy photon reactions ( $< 2$  MeV) and to the lack of suitable x-ray devices committed to such studies.

Since 1999, there have appeared additional positive reports [15-18] of triggered gamma emission from  $^{178m2}\text{Hf}$  and a report of a null measurement [25]. Clearly the question remains open whether low-energy triggering ( $< 100$  keV) can be accomplished for  $^{178m2}\text{Hf}$ .

The need for improved experiments to test triggering of  $^{178m2}\text{Hf}$ , to extend the understanding of triggering for  $^{180m}\text{Ta}$ , and to establish a closer connection between these studies and traditional nuclear physics has motivated the work conducted under this grant. This work has been considerable and resulted in fifteen publications, many of which have contributed significantly to the foundations of the field. The following provides a brief guide to these accomplishments, which are detailed in the reprints included in Appendix A.

### **PUBLICATIONS FOR WORK CONDUCTED UNDER GRANT<sup>3</sup>**

#### **Development of improved systematics for photonuclear reactions**

"Light-mass yields and fine structure of mass distributions in  $^{232}\text{Th}$  photofission,"  
S. A. Karamian, J. Adam, A. G. Belov, J. J. Carroll, Yu. V. Narseev,  
V. I. Stegailov and P. Chaloun, Phys. Rev. C **62**, 024601-1 (2000).

#### **Summary:**

Experiments at the Joint Institute for Nuclear Research, Dubna, Russia, were conducted to investigate fine structure in the mass distributions of fragments produced by photo-fission of  $^{232}\text{Th}$ . Bremsstrahlung endpoints of 7.5, 12 and 24 MeV were used.

<sup>3</sup> Seven published during grant period, eight submitted during, but published after, the grant period.



Main points:

1. Twenty-seven fission fragments were identified by post-irradiation gamma-ray spectroscopy.
2. Measured relative probabilities for the symmetric-fission and fine-structure amplitudes were compared with previous experiments.
3. The relevance of measured light nuclide yields was discussed as providing insight into possible ternary fission mechanisms.

"The new photoactivation facility at the 4.3 MV Stuttgart DYNAMITRON: setup, performance, and first applications," D. Belic, J. Besserer, C. Arlandini, J. de Boer, J. J. Carroll, J. Enders, T. Hartmann, F. Kappeler, H. Kaiser, U. Kneissl, M. Loewe, H. Maser, P. Mohr, P. von Neumann-Cosel, A. Nord, H. H. Pitz, A. Richter, M. Schumann, S. Volz, A. Zilges, Nucl. Inst. Meth. A **463**, 26 (2001).

Summary:

Photoactivation of the well-known isomer  $^{115m}\text{In}$  was investigated using the high-current DC DYNAMITRON accelerator at the University of Stuttgart. This facility supports studies with greater sensitivity for the interaction of photons in the MeV range with nuclei, combining photoactivation and nuclear resonance scattering techniques.

Main points:

1. Photoactivation of  $^{115m}\text{In}$  was investigated using bremsstrahlung over a range of 0.8 – 4.3 MeV. Isomer yield was measured post-irradiation by gamma-ray spectroscopy and showed distinctive evidence of individual intermediate states.
2. Nuclear resonance scattering spectra were obtained in-beam and identified strong, fragmented dipole transitions concentrated near energies corresponding to the photoactivation intermediate states.
3. Integral cross sections for photoactivation and resonance scattering were extracted from the respective yields.

Characterization of triggering for  $^{180m}\text{Ta}$

"Photoactivation of  $^{180}\text{Ta}^m$  and its implications for the nucleosynthesis of nature's rarest naturally occurring isotope," D. Belic, C. Arlandini, J. Besserer, J. de Boer, J. J. Carroll, J. Enders, T. Hartmann, F. Kappeler, H. Kaiser, U. Kneissl, M. Loewe, H. J. Maier, H. Maser, P. Mohr, P. von Neumann-Cosel, A. Nord, H. H. Pitz, A. Richter, M. Schumann, S. Volz, and A. Zilges, Phys. Rev. Lett. **83**, 5242 (2000).

## Summary:

The origin of the naturally-occurring isomer  $^{180m}\text{Ta}$  remains a mystery, with s-process nucleosynthesis in red giants being the most likely mechanism. Previously, it was determined that strong intermediate states near 2.8 and 3.6 MeV existed which allowed a depopulation of the isomer. This process was a triggered energy release, with concomitant gamma emission, from an isomer – the first (and only) conclusive demonstration of this effect. The unique DYNAMITRON facility at the University of Stuttgart was used to further investigate this process and search for additional intermediate states that were beyond the sensitivity of earlier studies. The combination of detectors, sample (highly-enriched in the isomer) and the accelerator provided a 4,000 $\times$  improvement in experimental sensitivity. This work was recognized by the American Physical Society as one of the top stories in physics for the year 2000 (see Fig. 3).

## Main points:

1. The intermediate state previously located at 2.8 MeV was confirmed in photoactivation experiments using enriched and natural tantalum targets.
2. A new intermediate state was found at about 1 MeV. Its integral cross section was deduced from the measured isomer depletion and found to be significantly lower than that of the 2.8-MeV level. Nevertheless, its presence at an energy corresponding to considerably more incident photon flux required a reduction in the integral cross section assigned to the higher-lying intermediate state.
3. The impact of this new low-lying intermediate state on s-process nucleosynthesis was discussed. The stellar photon bath corresponding to the standard s-process model would be expected to provide sufficient depletion of the isomer that none

## Physics News In 2000

**THE RAREST NATURALLY OCCURRING ISOTOPE**, tantalum-180, is less of a rarity now. The isotope is scarce because its nucleosynthesis is mainly bypassed in the processes that produced most of the heavy elements we find here on Earth: the s-process (slow neutron capture within stars) and the r-process (rapid neutron capture during supernova explosions). The  $^{180}\text{Ta}$  nucleus, with a half-life of more than  $10^{15}$  years, is the only naturally occurring nuclear isomer; it is essentially in a perpetually excited state. Now, a group of physicists in Germany has found that some  $^{180}\text{Ta}$  can arise in the s-process. At the Dynamitron accelerator in Stuttgart, they exposed this isotope to an intense beam of gamma rays, simulating the thermal-photon conditions inside a star, and found that the long-lived isomer can be jarred through an intermediate state into a short-lived ground state (with an effective half-life of only a month,  $10^{17}$  times less than that of the isomer!). Moreover, the temperature of the radiation field corresponds to that of the brief "helium flash" phase in a star's evolution; rapid convection of the star's material would quickly remove the  $^{180}\text{Ta}$  to cooler regions, where it could then survive in its stable isomeric form. (D. Belic et al., *Phys. Rev. Lett.* **83**, 5242, 1999.)

Figure 3: Excerpt from the APS News insert on Physics News in 2000.

would survive. More detailed models of s-process environments, including different ( $\alpha$ , n) sources and mixing between hotter and colder regions, remain consistent with survival of  $^{180\text{m}}\text{Ta}$ .

"Photon cross sections for resonant de-excitation of nuclear isomers as a precursor to a gamma-ray laser," S. A. Karamian and J. J. Carroll, *Laser Phys.* **11**, 23 (2001).

#### Summary:

Photoactivation of isomers and related depletion (or triggering) reactions are characterized by integral rather than simple cross sections due to the broad continuum nature of typical irradiating photon fluxes. A consistent mathematical model of this integral cross section was needed in order to determine fundamental nuclear parameters, such as the well-known B values. This model was applied to the case of  $^{180\text{m}}\text{Ta}$  depletion (triggering), in light of recent improved measurements of intermediate states. The large value of integral cross section found in previous studies at 2.8 MeV was of concern as being larger than systematic Recommended Upper Limits and this issue was investigated.

#### Main points:

1. The Blatt-Weisskopf and Breit-Wigner mathematical models were compared as applied to reactions involving resonant photon absorption (for photoactivation and/or triggering). The connection between these equivalent formulations and B values for nuclear electromagnetic transitions was given.
2. The integral cross section of the 2.8-MeV intermediate state in  $^{180}\text{Ta}$  was examined using this formulation and in light of the discovery of the lower-lying intermediate state near 1 MeV. The improved value for the integral cross section of the higher-lying intermediate state was found to be in general agreement with systematics of the Recommended Upper Limits.

"Interpretation of the excitation and decay of  $^{180}\text{Ta}^{\text{m}}$  through a  $K^{\pi} = 5^{+}$  band," P. M. Walker, G. D. Dracoulis, and J. J. Carroll, *Phys. Rev. C* **64**, 061302(R) (2001).

#### Summary:

Recent improved measurements identified a number of new intermediate states that allow depopulation (triggering) of  $^{180\text{m}}\text{Ta}$ .<sup>4</sup> For the first time, it was possible to make identifications between the intermediate states for isomer depletion (triggering) with known levels obtained from independent spectroscopic studies.

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<sup>4</sup> By agreement with the authors of the next paper (through the Investigator), data was made available for this interpretive work in 2001 prior to publication of the full experimental paper in 2002.

Main points:

1. The three lowest-lying intermediate states in  $^{180}\text{Ta}$  were identified as corresponding to E1 transitions to the  $8^+$ ,  $9^+$  and  $10^+$  members of the  $J^\pi = K^\pi = 5^+$  band.
2. Significant, but not complete, K mixing was suggested by the branching ratios deduced for these transitions.
3. A scheme was suggested for identification of higher-lying intermediate states, based on expected band structures.

“Photo-induced depopulation of the  $^{180}\text{Ta}^m$  isomer via low-lying intermediate states: Structure and astrophysical implications,” D. Belic, C. Arlandini, J. Besserer, J. de Boer, J. J. Carroll, J. Enders, T. Hartmann, F. Käppeler, H. Kaiser, U. Kneissl, E. Kolbe, K. Langanke, M. Loewe, H. J. Maier, H. Maser, P. Mohr, P. von Neumann-Cosel, A. Nord, H. H. Pitz, A. Richter, M. Schumann, F.-K. Thielemann, S. Volz, and A. Zilges, *Phys. Rev. C* **65**, 035801-1-13 (2002).

Summary:

This paper provided a complete description of experiments using the University of Stuttgart DYNAMITRON on the photon depletion (triggering) of  $^{180m}\text{Ta}$ .

Main points:

1. A total of eight intermediate states were identified for depletion of  $^{180m}\text{Ta}$  below the earlier level at 2.8 MeV. Integral cross sections for all these intermediate states were extracted.
2. The previously-known intermediate state at 2.8 MeV was confirmed, but its integral cross section was revised downward since earlier experiments were insensitive to the lower-lying levels and thereby over-estimated the role of the higher-lying state.
3. Nuclear resonance scattering experiments were attempted with the highly-enriched tantalum target, but were unsuccessful in observing strong dipole resonances.
4. Implications of these intermediate states to s- and v-process stellar nucleosynthesis were discussed.

**Tests of low-energy triggering for  $^{178m2}\text{Hf}$**

“Improved method for detection of x-ray triggered decay of the  $^{178}\text{Hf}^{m2}$  isomer using a multidetector array,” J. J. Carroll, S. A. Karamian, M. K. Boyle, Y. Kaneko, R. Toman, D. Fowler, M. Helba, H. Roberts and F. J. Agee, *Laser Physics* **11**, 6 (2001).

### Summary:

Previous studies of triggering for the 31-year isomer of  $^{178\text{m}2}\text{Hf}$  were plagued by the difficulty in separating gamma rays emitted by triggered events from those due to spontaneous decay. In Refs. [13-18] triggering was inferred from statistically small enhancements in the numbers of gamma rays emitted while samples were under irradiation, compared with the numbers emitted without irradiation. The gamma rays in question were part of the spontaneous decay cascade from the long-lived isomer. The inability to separate triggered vs. natural emission by these transitions left experiments in a vague state as viewed by the nuclear physics community. Here a new experimental approach was introduced by which it was possible to separate triggered and spontaneous-decay emissions. This technique was based on the premise that some, if not all, triggered events may produce gamma cascades that bypass the 4-s m1 isomer. In this case, triggered and spontaneous events would be differentiated based on the total energy emitted simultaneously in the cascade (the natural decay cascade is stalled by the 4-s isomer, so the total energy of each part of the cascade reaches only a little above 1 MeV, compared with the total 2.5 MeV stored by the m2 isomer).

### Main points:

1. The natural advantage afforded by the 4-s isomer in  $^{178}\text{Hf}$  was discussed.
2. The expectation of some high-multiplicity, high-sum-energy events (bypassing the 4-s isomer) was discussed in the context of other experiments.
3. The design of a modest-cost multi-detector system was discussed, with high-efficiency scintillators being used to provide sum-energy calorimetry in coincidence with a single Ge detector.
4. Sources of backgrounds were examined – essentially no background was expected in the region of data space corresponding to high-multiplicity triggered emission.

“Initial search for triggered gamma emission from  $^{178}\text{Hf}^{\text{m}2}$  using the YSU miniball array,”  
J. J. Carroll, J. Burnett, T. Drummond, J. Lepak, R. Propri, D. Smith,  
S. A. Karamian, J. Adam, F. Stedile and F. J. Agee, *Hyperfine Int.* **143**, 37 (2002).

### Summary:

A first test (late 2001) of triggering for  $^{178\text{m}2}\text{Hf}$  was performed using the YSU miniball multi-detector system. A target containing isomeric  $^{178}\text{Hf}$  was irradiated with bremsstrahlung from a medical radiographic x-ray tube and coincident bi-dimensional data (Ge vs. scintillator sum energy) were obtained.

### Main points:

1. Background was found to be zero in the region of data space corresponding to high-multiplicity triggered events.
2. No signal consistent with triggered gamma emission was observed. An upper limit of  $10^{-20} \text{ cm}^2 \text{ keV}$  for the integral cross section for triggering was extracted.

This limit does not contradict reports of positive indications of triggering at the level of  $10^{-22}$  cm<sup>2</sup> keV.

3. Further refinements to the system to narrow the coincidence window between detectors and lengthened irradiation times were indicated. These would move the limit of sensitivity far below that corresponding to the reported integral cross section for triggering.

"Gamma spectroscopy of Hf-178m2 using synchrotron x-rays," H. E. Roberts, M. Helba, J. J. Carroll, J. Burnett, T. Drummond, J. Lepak, R. Propri, Z. Zhong and F. J. Agee, *Hyperfine Int.* **143**, 111 (2002).

#### Summary:

A study of triggering for <sup>178m2</sup>Hf was conducted during September 2001 using a single Ge detector at Brookhaven National Laboratory's National Synchrotron Light Source. A sample containing isomeric <sup>178</sup>Hf was irradiated with monochromatic radiation scanned over ranges covering the L<sub>1</sub>, L<sub>2</sub>, L<sub>3</sub> and 12.5-keV energies – triggering near the L<sub>1</sub> and L<sub>3</sub> edges had been claimed by other workers. The latter energy is that of the primary initial decay from the 31-year isomer.

#### Main points:

1. Monochromatic radiation having a width of about 2 eV was scanned in 2/3 eV steps across the energy ranges of interest. Beam intensities before and after the hafnium target were recorded by ionization chambers and these and gamma-ray spectra were obtained during 20-s intervals at each monochromatic beam energy. Photon intensity within the 2-eV bandwidth was about  $4.5 \times 10^9$  photons/(s mm<sup>2</sup>).
2. Bi-dimensional data sets showing gamma-ray spectra vs. monochromatic beam energy were examined visually for evidence of modified or new gamma lines. None were observed.
3. The data sets were scanned by computer to determine if statistically-significant variations in peaks (or backgrounds) were present. No significant variations were detected.
4. An upper limit on the integral cross section for triggering of <sup>178m2</sup>Hf was extracted, having a value of  $10^{-25}$  cm<sup>2</sup> keV.

#### Examination of candidate isomers and trigger approaches

"X-ray driven gamma emission," J. J. Carroll, S. A. Karamian, L. A. Rivlin and A. A. Zadernovsky, *Hyperfine Int.* **135**, 3 – 50 (2001).

#### Summary:

This paper reviewed the field of study related to triggered gamma emission, driven by incident photons. It comprised the most complete overview to-date of experimental and theoretical considerations in quantum nucleonics.

Main points:

1. The experimental results for depletion of  $^{180m}\text{Ta}$  and systematics of related photonuclear reactions was discussed.
2. General experimental methods for the detection of triggered gamma emission were described.
3. A model was described for the creation of a photon chain reaction in a hot plasma.
4. Mathematical foundations were described for stimulated emission in nuclear populations as a basis for a gamma-ray laser.
5. A model for a gamma-ray laser based on recoil was reviewed.

"On the way to x-ray driven triggering of nuclear isomers," S. A. Karamian and J. J. Carroll, *Laser Phys.* **12**, 310 – 313 (2002).

Summary:

This paper considered means by which the probability of triggering might be enhanced through mechanisms such as NEET, perhaps in hot plasma environments.

Main points:

1. The state-of-the-art in trigger and related photonuclear reaction studies was reviewed. The general systematics of these reactions was discussed.
2. The NEET process was investigated in general as a means of enhancing trigger cross sections in hot plasmas.
3. Specific candidates for study were introduced and a parameter study presented.

"Non-radiative triggering of long-lived nuclear isomers," A. A. Zadernovsky and J. J. Carroll, *Hyperfine Int.* **143**, 153 (2002).

Summary:

Attention has been recently focused on the concept of using mechanisms not relying on real photons to cause triggered gamma emission from isomeric nuclei. This paper considered various such mechanisms, including Coulomb excitation and NEET.

Main points:

1. Mechanisms of Coulomb excitation, NEET and NEEC were presented.
2. Projected cross sections were obtained for these processes.
3. A parameter study was presented for several candidate isomers.



### Study of approaches to a gamma-ray laser

"Proposal for Observation of a Hidden Nuclear Population Inversion," F. J. Agee, J. J. Carroll, L. A. Rivlin and V. Vuletic, *Hyperfine Int.* **143**, 7 (2002).

#### Summary:

Approaches to the development of a gamma-ray laser have been frustrated by technological problems for the past forty years, since the first suggestion of such a device in 1961. One of the main requirements of any scheme is that the natural linewidth of a potential nuclear lasing transition be maintained, i. e. without broadening due to Doppler shifts, etc. For this reason, attention has largely concentrated on recoilless Mössbauer emission in crystals. This paper re-examines the concept of recoil-assisted gamma-ray lasing in which a cooled atomic population provides the narrow linewidth. Recoil of pumped (pre-lasing) nuclei produces a sub-population of inverted nuclei (a "hidden" inversion).

#### Main points:

1. General requirements for a gamma-ray laser were discussed.
2. The approach of recoil-assisted gamma-ray lasing was described.
3. The impact of recent and promising new techniques of laser cooling on this approach was discussed and a parameter study presented.
4. A proposed experiment was discussed by which to observe the presence of a hidden nuclear population inversion.

"Possibility of combining nuclear level pumping in a plasma with lasing in a solid," S. A. Karamian and J. J. Carroll, *Hyperfine Int.* **143**, 69 (2002).

#### Summary:

Technological challenges to a solid-state gamma-ray laser represent the contradiction between the needs for strong pumping of nuclear levels suitable for lasing and for maintenance of recoil-free (Mössbauer) emission. Here an approach was presented that may by-pass this difficulty by allowing pumping within a plasma environment and lasing in an ensemble which rapidly cools during deposition on a solid substrate.

#### Main points:

1. The requirements for gamma-ray lasing were described.
2. Pumping of a population inversion was analyzed for nuclei exposed to the radiation bath within a hot plasma created by a high-power laser.
3. Time scales of required nuclear level lifetime and cooling and deposition processes were compared. A parameter study was presented for candidate nuclei.

## Study of isomer production mechanisms

"Beam-based production of  $^{178m2}\text{Hf}$ ," J. Paul Farrell, V. Dudnikov, J. J. Carroll and G. Merkel, *Hyperfine Int.* **143**, 55 (2002).

### Summary:

Nuclear reactions capable of producing the 31-year isomer of  $^{178}\text{Hf}$  have recently become of more interest due to the potential applications of this metastable state. A number of studies have been conducted on neutron-induced reactions, including at the Joint Institute for Nuclear Research. This paper described experiments performed to measure particle-beam production of this isomer by incomplete fusion reactions, with the hopes of higher relative yield compared to ground-state  $^{178}\text{Hf}$  production.

### Main points:

1. An experiment was described to measure the cross section for the  $^{176}\text{Yb}(^9\text{Be}, \alpha 3n)^{178m2}\text{Hf}$  incomplete fusion reaction.
2. The cross section was extracted and found to be on the order of a few millibarns.

## EDUCATIONAL IMPACT

Although beyond the typical level of research conducted at comprehensive undergraduate institutions like Youngstown State University, this project has benefited tremendously from the involvement of a number of students at the sophomore, junior and senior levels, and from several different departments. Students Mary Kate Boyle, Yuki Kaneko, Joel Lepak and Rick Toman contributed to every aspect of the planning, design and construction of detector systems and benefited greatly from the unique experience on this project. Ms. Boyle completed a M.S. degree in physics and is now gainfully employed in the private sector on missile defense, while Ms. Kaneko is completing a Ph.D. in astronomy. Mr. Lepak is seeking a Ph.D. in mathematics. Mr. Toman is gainfully employed in the private sector, involved in the manufacture of laser crystals for inertial confinement fusion devices, in collaboration with workers at Lawrence Livermore National Laboratory.

The impact of this project on the education of young scientists has already been considerable and will only expand in the future.

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## APPENDIX A

### PUBLICATIONS FROM WORK CONDUCTED DURING GRANT PERIOD

- "Light-mass yields and fine structure of mass distributions in  $^{232}\text{Th}$  photofission," S. A. Karamian, J. Adam, A. G. Belov, J. J. Carroll, Yu. V. Noursev, V. I. Stegailov and P. Chaloun, *Phys. Rev. C* **62**, 024601-1 (2000). **REPRINT**
- "The new photoactivation facility at the 4.3 MV Stuttgart DYNAMITRON: setup, performance, and first applications," D. Belic, J. Besserer, C. Arlandini, J. de Boer, J. J. Carroll, J. Enders, T. Hartmann, F. Kappeler, H. Kaiser, U. Kneissl, M. Loewe, H. Maser, P. Mohr, P. von Neumann-Cosel, A. Nord, H. H. Pitz, A. Richter, M. Schumann, S. Volz, A. Zilges, *Nucl. Inst. Meth. A* **463**, 26 (2001). **REPRINT**
- "Photoactivation of  $^{180}\text{Ta}^m$  and its implications for the nucleosynthesis of nature's rarest naturally occurring isotope," D. Belic, C. Arlandini, J. Besserer, J. de Boer, J. J. Carroll, J. Enders, T. Hartmann, F. Kappeler, H. Kaiser, U. Kneissl, M. Loewe, H. J. Maier, H. Maser, P. Mohr, P. von Neumann-Cosel, A. Nord, H. H. Pitz, A. Richter, M. Schumann, S. Volz, and A. Zilges, *Phys. Rev. Lett.* **83**, 5242 (2000). **REPRINT**
- "Photon cross sections for resonant de-excitation of nuclear isomers as a precursor to a gamma-ray laser," S. A. Karamian and J. J. Carroll, *Laser Phys.* **11**, 23 (2001). **REPRINT**
- "Interpretation of the excitation and decay of  $^{180}\text{Ta}^m$  through a  $K^\pi = 5^+$  band," P. M. Walker, G. D. Dracoulis, and J. J. Carroll, *Phys. Rev. C* **64**, 061302(R) (2001). **REPRINT**
- "Photo-induced depopulation of the  $^{180}\text{Ta}^m$  isomer via low-lying intermediate states: Structure and astrophysical implications," D. Belic, C. Arlandini, J. Besserer, J. de Boer, J. J. Carroll, J. Enders, T. Hartmann, F. Kappeler, H. Kaiser, U. Kneissl, E. Kolbe, K. Langanke, M. Loewe, H. J. Maier, H. Maser, P. Mohr, P. von Neumann-Cosel, A. Nord, H. H. Pitz, A. Richter, M. Schumann, F.-K. Thielemann, S. Volz, and A. Zilges, *Phys. Rev. C* **65**, 035801-1-13 (2002). **REPRINT**
- "Improved method for detection of x-ray triggered decay of the  $^{178}\text{Hf}^{m2}$  isomer using a multidetector array," J. J. Carroll, S. A. Karamian, M. K. Boyle, Y. Kaneko, R. Toman, D. Fowler, M. Helba, H. Roberts and F. J. Agee, *Laser Physics* **11**, 6 (2001). **REPRINT**
- "Initial search for triggered gamma emission from  $^{178}\text{Hf}^{m2}$  using the YSU miniball array," J. J. Carroll, J. Burnett, T. Drummond, J. Lepak, R. Propri, D. Smith, S. A. Karamian, J. Adam, F. Stedile and F. J. Agee, *Hyperfine Int.* **143**, 37 (2002). **REPRINT**

- "Gamma spectroscopy of  $\text{Hf}178\text{m}2$  using synchrotron x-rays," H. E. Roberts, M. Helba, J. J. Carroll, J. Burnett, T. Drummond, J. Lepak, R. Propri, Z. Zhong and F. J. Agee, *Hyperfine Int.* **143**, 111 (2002). **REPRINT**
- "X-ray driven gamma emission," J. J. Carroll, S. A. Karamian, L. A. Rivlin and A. A. Zadernovsky, *Hyperfine Int.* **135**, 3 – 50 (2001). **REPRINT**
- "On the way to x-ray driven triggering of nuclear isomers," S. A. Karamian and J. J. Carroll, *Laser Phys.* **12**, 310 – 313 (2002). **REPRINT**
- "Non-radiative triggering of long-lived nuclear isomers," A. A. Zadernovsky and J. J. Carroll, *Hyperfine Int.* **143**, 153 (2002). **REPRINT**
- "Proposal for Observation of a Hidden Nuclear Population Inversion," F. J. Agee, J. J. Carroll, L. A. Rivlin and V. Vuletic, *Hyperfine Int.* **143**, 7 (2002). **REPRINT**
- "Possibility of combining nuclear level pumping in a plasma with lasing in a solid," S. A. Karamian and J. J. Carroll, *Hyperfine Int.* **143**, 69 (2002). **REPRINT**
- "Beam-based production of  $^{178\text{m}2}\text{Hf}$ ," J. Paul Farrell, V. Dudnikov, J. J. Carroll and G. Merkel, *Hyperfine Int.* **143**, 55 (2002). **REPRINT**